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13. ABSTRACT (Maximum 200 words)

Experiments on epitaxial Ni films grown on epitaxial Cu films grown on single crystal (100) Si wafers in a molecular beam epitaxy system modified to make in situ magneto-optic Kerr effect (MOKE) measurements were carried out. Ex situ transmission electron microscopy and wafer curvature measurements were made and it was shown that the onset of misfit accommodation, the decrease in Ni strain with increasing film thickness, and the increase in misfit dislocation density were consistent with existing models for misfit accommodation In situ MOKE studies and ex situ vibrating sample magnetometry studies of the magnetic consequences of the changing strain with Ni film thickness showed that the magnetic easy axis of the Ni/Cu(001) films was perpendicular to the film plane over the range 1.5 to approximately 6 nm. We have found from these studies that the low-thickness transition from perpendicular to in-plane magnetization with decreasing film thickness (seen also in ultra-thin Fe/Cu and Fe/Ag) is NOT associated with the critical thickness leading to the onset of MD formation, but results instead from a complex interplay of magnetostatic and magnetoelastic energies, and of the Néel magnetic surface anisotropy as well as the surface magneto-elastic anisotropy.

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Anisotropy in Epitaxial Films

Final Report

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The objective of the program was to use the magneto-optic Kerr effect (MOKE) as an *in-situ* probe for monitoring and characterizing the magnetic anisotropy associated with misfit dislocations (MDs) which form during growth of epitaxial Ni films.

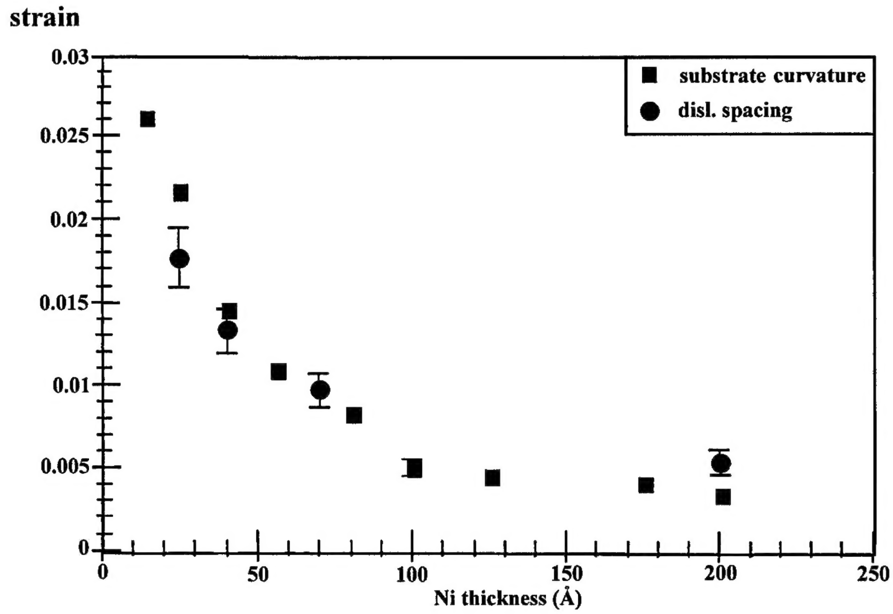

Figure 1: Strain as indicated by measured misfit dislocation spacings and by substrate curvature for epitaxial Ni films of different thickness, grown on epitaxial Cu films grown epitaxially on (001) single crystal Si wafers using molecular beam epitaxy.

For Ni on Cu(001) thermodynamic theory predicts a critical thickness of h_c=1.8nm for formation of MDs (2.6% lattice mismatch). Above h_c, the MD spacing and residual strain decrease approximately as h^{-1}. We carried out experiments on epitaxial Ni films grown on epitaxial Cu films grown on single crystal (100) Si wafers in a Molecular Beam Epitaxy (MBE) system which we modified to make in situ MOKE measurements. We characterized misfit accommodation through extensive *ex situ* transmission electron microscopy (TEM), in which we observed the onset of misfit accommodation through MD formation for Ni films thicker than about 2.5 nm.[1,2] We measured the MD density as a function of Ni film thickness, and made ex situ strain measurements as a function of film thickness using TEM Moiré fringe analysis and optical interferometry for substrate curvature measurements[1-4]. As expected, we found that the misfit was accommodated by dislocations lying along [110] and [110] of Cu on [100] and [101] of Si directions. However, we also found that while some of the MD's had Burgers vectors lying in slip planes and inclined at 60^0 to the line direction, we found that much of the misfit was accommodated by '90^0' dislocations with Burgers vectors in the (001) plane, the plane of the Ni/Cu interface. Mechanisms for the formation of these dislocations were described, and it was shown that our observations of the onset of misfit accommodation, the decrease in Ni strain with increasing thickness, and the increase in misfit dislocation density with increasing film thickness (Figure 2) were consistent with existing models for misfit accommodation[1-4].

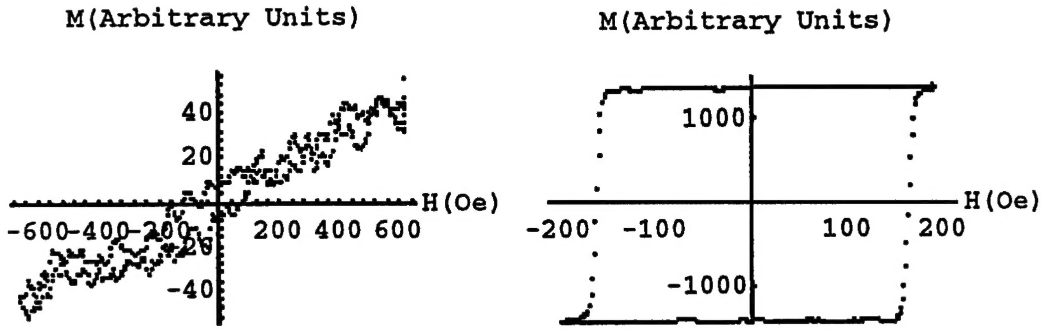

Figure 2: Longitudinal (left) and polar (right) M-H loops for 35 Å Ni / 3000 Å Cu/Si(100).

We also found that as the strain changed due to film thickening, the shape of the M-H loop (which is determined by magnetic anisotropy) varied in a predictable way because of the strong magnet-elastic coupling in Ni, the strong magnetic energy of the Cu-Ni interface, and the magnetostatic energy (Fig. 3).[2, 5-6]

MOKE studies of the magnetic consequences of the changing strain showed that the magnetic easy axis of the Ni/Cu(0010) films is perpendicular to the film plane over the range 1.5 to approximately 6 nm. Ni films deposited on CuNi substrates for which the misfit was smaller showed a smaller thickness range for perpendicular magnetization (Fig. 3).[6]

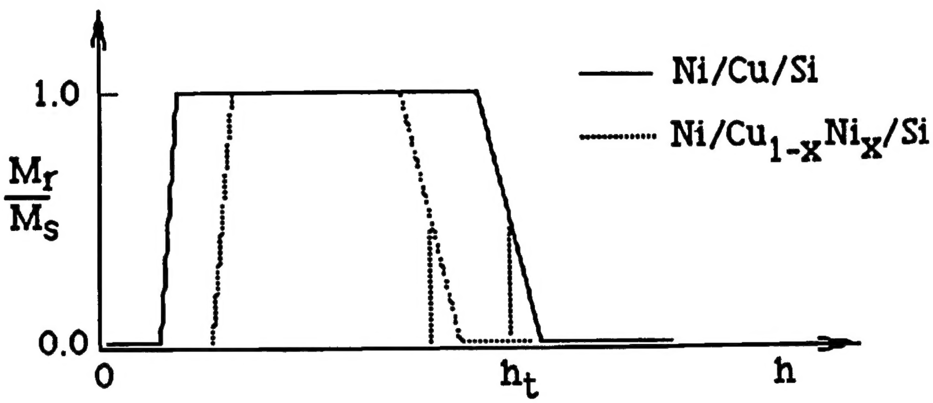

Figure 3: Variation to the perpendicular remanence normalized to the saturation magnetization vs. Ni film thickness in Ni/3000 Å Cu/Si (001) and in $Ni/Cu_{1-x}Ni_x/Si$ (001) for which the misfit strain is reduced.

In iron films the upper limit to the range of perpendicular magnetization is approximately 0.4 nm. This sharp difference between Fe and Ni films is due largely to the much smaller magnetostatic energy of Ni. Nevertheless, this difference also points to strong effects in the Ni films favoring perpendicular magnetization. We found that the film strain plays and important role in the perpendicular magnetization of Ni.

The important contributions to the anisotropy energy are usually identified[7] as: magnetostatic $f_{MS} = -2\pi M_s^2 \sin^2\theta$, magnetoelastic $f_{ME} = 2B_1 e_0 \sin^2\theta$, and the Néel magnetic surface anisotropy $f_N = K^S \sin^2\theta$. Here B_1 is the ME coupling coefficient and K_S is the magnetic surface anisotropy of the film. The magnetocrystalline anisotropy energy density of Ni is small compared to the other contributions and is ignored. The total magnetic anisotropy energy density can then be expressed as:

$$f_M = K^{eff} \sin^2\theta, \tag{1}$$

$$K^{eff} = 2B_1 e_o - 2\pi M_S^2 + 2K^s / h. \tag{2}$$

Here M_s is the saturation magnetization and θ is the angle that the magnetization vector makes with the film normal, f_{MS} is of order 1.5 x 10^6 erg/cm^3, h is the film thickness, and above the critical thickness e_o goes approximately as $\eta h_c/h$. The convention here is the $K^{eff} > 0$ favors a perpendicular magnetization whereas $K^{eff} < 0$ favors and in-plane magnetization. The magnetoelastic term requires the film strain as input.

Careful studies of Cu/Ni/Cu/Si(001) sandwiches showed that these terms alone are not enough to fit the measured K^{eff} data. This can be seen by plotting Eq. 2 as $(K^{eff} + 2\pi M_s)h = 2B_1\eta h_c + 2K^s$, which is a constant (dashed line, Fig. 4). It must be recognized[7,8] that the Néel model requires a surface ME term, $B^s e(h)/h$, as well as a surface anisotropy term. When this term is included, a much better fit to the anisotropy data is obtained (solid line, Fig. 4).

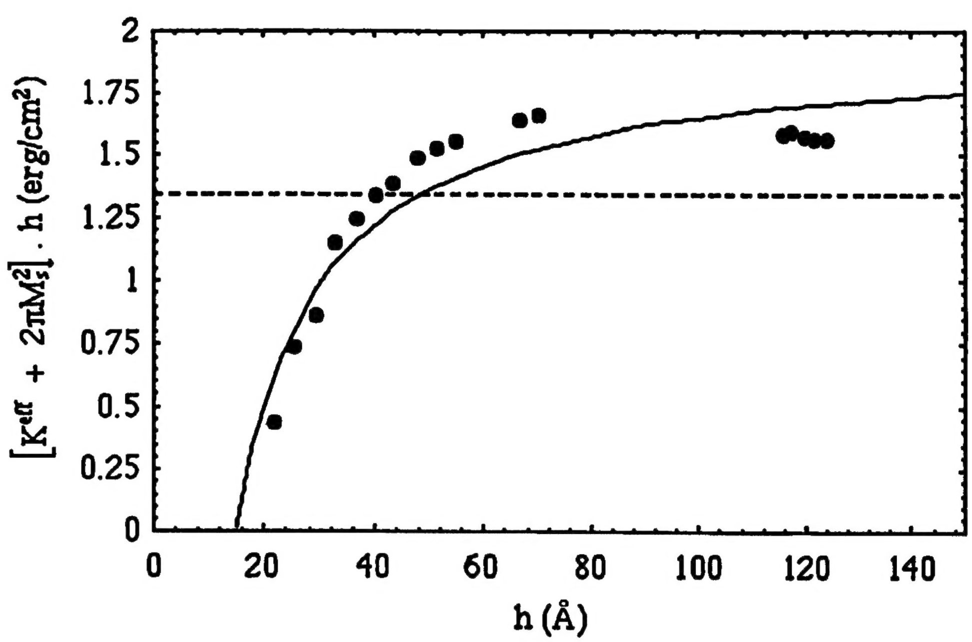

Figure 4: $[K^{eff} = 2\pi M_S^2]$. h versus h for the data of Jungblut *et al* [J. Appl. Phys. **75**, p.6424, 1994] on Cu/Ni/Cu(001) sandwiches, compared to Equation 2 (the dashed line) and compared to Equation 2 with a magnetoelastic surface anisotropy term, $B^s e(h)/h$

Our work was the first study of epitaxial Ni/Cu to combine independent measures of strain, misfit dislocation structure, and magnetic properties. We were the first to report and quantitatively account for the thickness dependence of the bulk and surface magnetoelastic terms in epitaxial Ni films[8].

We have found from these studies that the low-thickness transition from perpendicular to in-plane magnetization with decreasing film thickness (seen also in ultra-thin Fe/Cu and Fe/Ag) is NOT associated with the critical thickness for the onset of MD formation. Rather it is a result of the complex interplay of the energies in Equation 2, modified to include surface magneto-elastic anisotropy. Certainly the variation of film strain associated with the MDs contributes to the variation of anisotropy with film thickness. More importantly we have shown that the return to in-plane magnetization in the ultra-thin film limit is due to the negative sign of the magnetoelastic surface energy.

We have also demonstrated in this study that the surface energy of the Cu/Ni interface is strongly positive rather than negative as concluded earlier by Jungblut et al. There is no other explanation for the approximate doubling of the Ni thickness range over which perpendicular magnetization is observed when the number of Cu/Ni interfaces goes from one (in Ni/Cu/Si(001)) to two (in Cu/Ni/Cu/Si(001)).

Finally, these observations have been confirmed and their implications shown clearly with magnetic force microscopy images. This work, conducted with collaborators at the University of B. Gasel, Switzerland after the completion of the ARO program was published in Phys. Rev. Letters[9] and has been the subject of invited talks at the 1995 MMM conference[10], the March 1996 APS Meeting and the October 1996 AVS meeting.

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Bochi, G., Ballentine, C.A., Inglefield, H.E., Thompson, C.V., O'Handley, R.C., *Perpendicular Magnetic Anisotropy, Domains, and Misfit Strain in Epitaxial* $Ni/Cu_{1-x}Ni_x/Si$ *(001) Thin Films*, Physical Review B **52(10)**, pp. 7311, 1995.

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Degrees granted:

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